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Binding Energy and Effective Mass of Charged Particles Trapped on Surfaces^{*}

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Abstract

We discuss the interaction of a charged particle with the surface excitations of a semi-infinite medium. We consider only the case where the particle is outside the material and trapped near the surface by its image potential. By the use of a cononical transformation which exhibits the bound states due to the image charge in the non-perturbed transformed Hamiltonian, we determine the binding energy and the effective mass of a charged particle for motion parallel to the surface. For the highest perpendicular sub-bands the corrections to the energy and mass of the particle are negligible. We have applied the method to determine the binding energy of the ground-state of an electron trapped near the LiF surface. We find the value $E(0) = -0.25$ eV.

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I. INTRODUCTION

Both theory and experiment have been extensively developed by treating the interaction of charged particles with collective excitations on surface media; for example, surface plasmons in metals and surface optical phonons in dielectric crystals.¹ This interaction with surface modes can occur when the particle is either inside or outside the surface. We consider here the case where the charged particle lies outside the material, interacting then only with surface modes. When the particle is inside the medium it is also necessary to treat the interactions with the bulk modes.

It is well known that one charged particle outside the material induces charges of opposite sign on the surface and is attracted to it by the image potential. It can form a bound state in which it is trapped near the surface of the material by the attractive image charge. It is also well known that the image potential is due to the surface modes of oscillations. These modes, when polarized, create electric fields which act back upon the electric charge. This is the source of the image potential.

Cole and Cohen,² and Cole³ have showed that surface states induced by the electrostatic image potential at insulators such as He, Ne, H₂ and D₂, both solid and liquid, have too small binding energies. In these weakly bound states, the electron is localized well away from the surface.

In this work we investigate the possibility that a charged particle may be trapped near the surface of a semi-infinite material by the charge-surface excitation interaction. The material may, in principle, be either solid or liquid. Our major purpose is to determine the binding energy and the effective mass of the ground state of a charge particle placed above the surface of a material.

We shall assume that the difference between the vacuum level and the bottom of the conduction band is much larger than the binding energy. In this situation we may simplify the problem by considering that the charge cannot penetrate the material. This approximation is discussed in detail both in reference (3) and in a recent review paper by Crandall⁴ for the case of electrons trapped near the surface of He⁴. It is probably also applicable for LiF where the potential barrier is much larger than the binding energy.⁵ In these cases, the electron wavefunction is concentrated in a region far from the surface and the energy required for the electron to penetrate the material is too high. We also assume

that the interaction between the charged particle and the surface excitations is very weak, which means that the particle will be loosely bound to the surface.

We proceed in a similar fashion to the canonical transformation technique employed by Freitas and Lobo.⁶ As it was pointed out in reference (6), the canonical transformation chosen has the merit of exhibiting the bound states due to the image potential in the unperturbed part of the transformed Hamiltonian.

In Section II we describe the model to be employed and we discuss the interaction between a charged particle and longwavelength excitation modes in a semi-infinite material. We show in the framework of classical electrodynamics that a charged particle placed outside the material can excite only surface modes. We employ a canonical transformation technique for the whole Hamiltonian of the system and, in the limit of weak-coupling constant, part of the kinetic energy of the particle is treated as a perturbation.

After this examination of the interaction of the charged particle with the surface modes, we present in Section III the results for the energy shift which arises in the second-order of perturbation theory. We calculate the binding energy of the ground-state of a particle trapped near the surface as a function of α_s , the coupling-constant, and also the effective mass of the particle, for motion parallel to the surface.

II. MODEL HAMILTONIAN

Consider a charged particle placed a distance $z > 0$ from the surface of a semi-infinite material. We take the material to fill the half-space $z \leq 0$. We assume that the dielectric constant $\epsilon(\omega)$ is frequency-dependent only. A more general description would have the dielectric constant frequency and wave-vector dependent. The approximation of neglecting the wave-vector dependence gives equations which are much easier to solve although it has no justification in all physical situations.

For the sake of clarity and completeness, we shall show, in a standard procedure, that in the longwavelength limit an external charged particle does not interact with bulk modes.^{7,8} The reason is that the fields associated with bulk modes are confined to the material only. The interaction between an electric charge e and a given collective mode is given by $e\phi(\vec{r})$, where $\phi(\vec{r})$ is the electrostatic potential associated with this oscillation. Since there is translational invariance along a direction parallel to the surface, the type of solutions we seek correspond to waves for which

$$\phi(\vec{r}, t) = \varphi(z) e^{i\vec{Q} \cdot \vec{R}} e^{-i\omega t}, \quad (2.1)$$

where \vec{R} is the projection of the position vector \vec{r} onto a plane parallel to the surface and \vec{Q} is a two dimensional wave-vector.

Assuming that there is no charge density or currents in this system, we have from Maxwell's equations

$$\epsilon(\omega) \left\{ \frac{d^2 \varphi}{dz^2}(z) - Q^2 \varphi(z) \right\} = 0, \quad z < 0 \quad (2.2)$$

and

$$\frac{d^2 \varphi}{dz^2}(z) - Q^2 \varphi(z) = 0, \quad z > 0. \quad (2.3)$$

The solutions of Eq. (2.2) are either $\epsilon(\omega) = 0$, or a linear combination of the two independent solutions, e^{Qz} and e^{-Qz} . The solution $\epsilon(\omega) = 0$ gives

the frequency of the bulk excitations and the second possibility describes the surface modes. Outside the material $\varphi(z)$ satisfies Eq. (2.3) and is given by

$$\varphi(z) = \varphi_1 e^{-Qz} \quad (2.4)$$

and for the bulk modes solution, $\varphi(z)$ assumes the form

$$\varphi(z) = \varphi_2 \sin(q_z z) + \varphi_3 \cos(q_z z), \quad (2.5)$$

where φ_1 , φ_2 and φ_3 are constants to be determined.

The boundary conditions at the surface, $z = 0$, say that the normal components of \vec{D} and the tangential components of \vec{E} must be continuous. Since $\epsilon(\omega) = 0$, we have $D_z = 0$ inside the material. Then, just outside $D_z = 0$, which requires from Eq. (2.4), $\varphi_1 \equiv 0$. So, the electrostatic potential $\phi(\vec{r})$ is identically zero outside the medium which means that the parallel electric field is zero just outside. The continuity of the parallel components of \vec{E} at $z = 0$ requires from Eq. (2.5), $\varphi_3 = 0$.

We conclude that the electrostatic fields associated with bulk oscillations are confined inside the material where the charge oscillations take place. These fields are derived from an electrostatic potential $\phi(\vec{r}, t)$, given by the following expression

$$\phi(\vec{r}, t) = \varphi_2 e^{i\vec{Q} \cdot \vec{R} - i\omega t} \sin(q_z z) \quad (2.6)$$

By examining the solutions of Eq. (2.2) which describe the surface modes and by matching at the surface, the fields inside and outside of the material, we obtain the condition

$$\epsilon(\omega) = -1$$

which determines the frequency of the surface oscillation modes.

Then the electrostatic fields associated with these modes are extended outside the material, an important distinction with bulk modes. In other words, a charged

particle outside the system can excite only surface modes.

In this work we shall discuss only the interaction of the low-energy charged particle with surface modes. By assuming, for simplicity, that the charge cannot enter the material, we may write the Hamiltonian of the system in three parts:

$$H = H_C + H_S + H_{int} \quad , \quad (2.7)$$

where H_C is the charged particle kinetic energy, H_S describes the surface modes in the absence of the particle and H_{int} is the interaction between the charged particle and the surface modes. The charged particle part has the form

$$H_C = - (\hbar^2/2m) \nabla^2 \quad , \quad (2.8)$$

where m is the mass of the particle. The surface modes part of the Hamiltonian can be written as

$$H_S = \hbar\omega_S \sum_{\vec{Q}} a_{\vec{Q}}^+ a_{\vec{Q}} \quad , \quad (2.9)$$

where the surface-wave frequency ω_S is given by the relation $\epsilon(\omega_S) = -1$, $a_{\vec{Q}}^+$ and $a_{\vec{Q}}$ are the creation and annihilation operators respectively for the surface excitations, obeying the usual commutation relations,

$$[a_{\vec{Q}}, a_{\vec{Q}'}^+] = \delta_{\vec{Q}\vec{Q}'} \quad , \quad (2.10a)$$

$$[a_{\vec{Q}}, a_{\vec{Q}'}] = [a_{\vec{Q}}^+, a_{\vec{Q}'}^+] = 0 \quad (2.10b)$$

The interaction between the particle and the surface modes is given by the expression¹:

$$H_{int} = 2\pi i \left(\frac{\hbar e^2 g}{2A\omega_S} \right)^{\frac{1}{2}} \sum_{\vec{Q}} \frac{e^{-Qz}}{\sqrt{Q}} \left(e^{-i\vec{Q} \cdot \vec{R}} a_{\vec{Q}}^+ - e^{i\vec{Q} \cdot \vec{R}} a_{\vec{Q}} \right) \quad , \quad (2.11)$$

where A is the area of the surface, z is the distance of the particle from the surface and the parameter β has different value for each type of excitation. For example, it will be

$$\beta = \omega_S^2 / 2\pi \quad \text{plasmons} \quad (2.12a)$$

$$\beta = \frac{\omega_S^2}{2\pi} \left(\frac{\epsilon_S - 1}{\epsilon_S + 1} - \frac{\epsilon_\infty - 1}{\epsilon_\infty + 1} \right) \quad \text{phonons,} \quad (2.12b)$$

with ϵ_S , the static dielectric constant of the material and ϵ_∞ , the optical dielectric constant.

In analogy with the bulk excitations, we define the coupling-constant α_S , as

$$\alpha_S = \frac{2\pi e^2 \beta}{\omega_S^2} \left(\frac{m}{2\hbar^3 \omega_S} \right)^{\frac{1}{2}} \quad (2.13)$$

In a manner similar to Freitas and Lobo,⁶ we introduce a canonical transformation S which removes the coordinate \vec{R} and gives a convenient zeroth order Hamiltonian with bound states for the external charged particle.

We define the unitary operator

$$S = \exp \left(\sum_{\vec{Q}} f_{\vec{Q}} a_{\vec{Q}}^+ - f_{\vec{Q}}^* a_{\vec{Q}} \right), \quad (2.14)$$

with

$$f_{\vec{Q}} = - (2\pi i) (e/\hbar\omega_S) (\hbar\beta/2A\omega_S)^{\frac{1}{2}} \frac{e^{-i\vec{Q}\cdot\vec{R}-Qz}}{\sqrt{Q}}. \quad (2.15)$$

The new Hamiltonian \mathcal{H} will be generated by application of the unitary transformation S in the standard manner:

$$\mathcal{H} = S^{-1} H S \quad (2.16)$$

The explicit form of \mathcal{H} after expanding up to second order over the coupling constant has the following expression:

$$\begin{aligned}
\mathcal{H} = & -\frac{\hbar^2}{2m} \nabla^2 + \sum_{\vec{Q}} \hbar \omega_S a_{\vec{Q}}^+ a_{\vec{Q}}^- \\
& - \sum_{\vec{Q}} \left(\frac{2\pi^2 \hbar^3 \omega_S}{m A^2} \right)^{\frac{1}{2}} \alpha_S \frac{e^{-Qz}}{Q} \\
& - \frac{\hbar^2}{2m} \left\{ \left[U, \nabla^2 \right] + \frac{1}{2}! \left[U, \left[U, \nabla^2 \right] \right] + \dots \right\}, \quad (2.17)
\end{aligned}$$

where

$$U = \sum_{\vec{Q}} \left(f_{\vec{Q}} a_{\vec{Q}}^+ - f_{\vec{Q}}^* a_{\vec{Q}}^- \right), \quad (2.18)$$

and $[,]$ is a commutator.

By performing the \vec{Q} summation on the third term of the right-hand side of Eq. (2.17) and by combining this result with the $\partial^2/\partial z^2$ term of ∇^2 , we get

$$\begin{aligned}
\mathcal{H} = & -\frac{\hbar^2}{2m} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) + \left(-\frac{\hbar^2}{2m} \frac{\partial^2}{\partial z^2} - \frac{\pi^2 \hbar^3}{2m \omega_S^2} \frac{e^2}{z} \right) \\
& + \sum_{\vec{Q}} \hbar \omega_S a_{\vec{Q}}^+ a_{\vec{Q}}^- - \frac{\hbar^2}{2m} \sum_{\vec{Q}} \left\{ \left[\left(f_{\vec{Q}} a_{\vec{Q}}^+ - f_{\vec{Q}}^* a_{\vec{Q}}^- \right), \nabla^2 \right] + \dots \right\}. \quad (2.19)
\end{aligned}$$

In the weak-coupling limit, first order in α_S , the terms which come from the commutator in Eq. (2.19) are treated as perturbations to the new unperturbed Hamiltonian, $\mathcal{H} = \mathcal{H}_C + \mathcal{H}_S$,

$$\mathcal{H}_C = \frac{\hbar^2}{2m} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) + \left(-\frac{\hbar^2}{2m} \frac{\partial^2}{\partial z^2} - \frac{\pi^2 \hbar^3 e^2}{2m \omega_S^2} \frac{1}{z} \right)$$

with the restriction $z > 0$, and

$$H_S = H_S = \sum_{\vec{Q}} \hbar \omega_S a_{\vec{Q}}^+ a_{\vec{Q}}$$

We write the unperturbed state vector corresponding to the total momentum \vec{K} parallel to the surface in the form

$$|\Psi\rangle = \frac{e^{i\vec{K} \cdot \vec{R}}}{2\pi} \chi_n(z) \pi_q |v_q\rangle, \quad (2.21)$$

where $\chi_n(z)$ are the normalized s-wavefunction of the one-dimensional hydrogen-like Hamiltonian,

$$\chi_n(z) = \frac{2z}{a^{3/2} n^2} \sqrt{\frac{(n-1)!}{(n!)^3}} \left(\exp \frac{-z}{na} \right) L_n^1 \left(\frac{2z}{na} \right), \quad (2.22)$$

where $a = (2\omega_S^2 \hbar^2 / m e^2 \beta)$ is the effective Bohr radius, L_n^1 is a modified Laguerre polynomial and $\pi_q |v_q\rangle$ is the wavefunction of the excitation.

In the next section we attempt to determine the two quantities of interest here, namely, the binding energy $E(0)$, and the effective mass m^* of the ground-state of a charged particle for motion parallel to the surface.

III. BINDING ENERGY AND EFFECTIVE MASS

It is a straightforward matter to calculate the energy of a charged particle for the wavefunction described in Eq. (2.21). Strictly speaking, we will determine the energy within the zero plus one excitation mode space and in the limit of weak-coupling constant.

In this approximation the eigensolutions of H_C are

$$E_n^0(\vec{K}) = \frac{\hbar^2 K^2}{2m} + E_n, \quad (3.1)$$

where

$$E_n = -\left(\frac{\pi e^2}{2\omega_s^2}\right)^2 \frac{m}{2\hbar^2} \frac{1}{n^2} \quad (3.2)$$

is the energy spectrum of the s-hydrogen-like atom. Eq. (3.1) describes a set of surface sub-bands with band index n just as in the semiconductor surface inversion layers problem.⁹

The first correction to the energy, $E_n^0(\vec{K})$ of the particle arises in the second order of perturbation theory. Thus, we have

$$E_n^{(2)}(\vec{K}) = \sum_{n', \vec{K}', \vec{p}} \frac{|\langle \vec{K}', n'; \dots 1_{\vec{p}} \dots | \left[\vec{F}_{\vec{p}}, H_C \right] a_{\vec{p}}^{\dagger} | \vec{K}, n; \dots 0_{\vec{p}} \dots \rangle|^2}{E_n^0(\vec{K}) - E_{n'}^0(\vec{K}') - \hbar\omega_s} \quad (3.3)$$

In the process described by this second-order calculation, the intermediate states correspond to a particle in state n' and \vec{K}' and to one surface excitation with wave-vector \vec{p} .

It is very difficult to get values of the correction to the energy and effective mass from this general expression. Therefore, we simplify the calculations by assuming that we are interested only in the case where the particle remains on the lowest perpendicular sub-band. This is the situation discussed by Crandall⁴ to explain the long life-time of an electron trapped near the surface of He^4 .

For this case we find

$$E_1^{(2)}(\vec{K}) = \left(\frac{2\pi}{\hbar\omega_S} \right)^2 \frac{64 e^{2\hbar\beta}}{2A\omega_S} \sum_{\vec{p}} \left(-\frac{\hbar^2 \vec{K}^2}{2m} + \frac{\hbar^2 \vec{p}^2}{2m} \right)^2 \frac{1}{p (2+\alpha p)^6} \times \frac{1}{\frac{\hbar^2}{m} \vec{K} \cdot \vec{p} - \frac{\hbar^2 \vec{p}^2}{2m} - \hbar\omega_S} \quad (3.4)$$

Let us remind the reader that the significant contribution to the sum in Eq. (3.4) comes from values $p \lesssim 1/r_C$, $r_C = (\hbar/2m\omega_S)^{1/2}$ a length the order of the radius of a bulk excitation, because of the exponential dependence of Eq. (2.11). On the other hand, for $a \ll 2r_C$, we may approximate $2+\alpha p \approx 2$. By replacing the sum by integrations according to

$$\sum_{\vec{p}} \rightarrow \frac{A}{(2\pi)^2} \int d^2 p,$$

we get the following value for $\Delta E(0)$,

$$\Delta E(0) = -0.119 \alpha_S \hbar\omega_S. \quad (3.5)$$

This result fits very well Evans and Mills' theory¹⁰ of surface polarons, provided by the weak-coupling and no penetration of the variational particle wavefunction into the material. As can be seen from Eq. (3.5) for small values of α_S , the particle is weakly bound to the surface which means that the binding energy is too small compared to the surface excitation energy $\hbar\omega_S$.

Strictly speaking, our results are valid only for α_S smaller than 1 but they fit quite well the results given in reference (10) for the binding energy for α_S as large as 4. The physical picture of this small binding energy can be understood in the following way: to localize the particle near the surface it costs a certain amount in kinetic energy. But, for small α_S , the decrease in energy due to the particle-surface oscillation modes interaction in clothing

the particle with the surface excitation is nearly cancelled by the cost in kinetic energy.

We also have estimated the correction to the ground-state binding energy which arises from the second perpendicular sub-band. However, we found that this contribution was too small.

We applied this weak-coupling limit method to estimate the binding energy of an electron trapped near an ionic crystal surface. For example, for L_1F the coupling-constant is $\alpha_S = 7.1$ and Eq. (3.4) gives for the binding energy of the ground-state the value

$$E(0) = -0.25 \text{ eV} ,$$

which is close to the result obtained by Sak.¹¹ Therefore, we feel that the weak-coupling limit is not realized for ionic crystals since the electron interacts very strongly with the surface oscillation modes.

We now report the effective mass results of the particle which is related to the coefficient of the quadratic term in the expanded energy, as

$$E(\vec{K}) = E(0) + \frac{\hbar^2 K^2}{2m^*} + \dots \quad (3.6)$$

Thus, by expanding Eq. (3.4) in terms of K , we get for m^* the expression

$$m^* = m \left(1 + \frac{1}{16} \pi \alpha_S \right) . \quad (3.7)$$

As can be compared, the correction to the mass of the particle is $\pi \alpha_S / 16$ whereas it is $\pi \alpha_S / 8$ for the weak-coupling constant theory of Sak.¹¹

As was pointed out in Sak's paper and also as can be seen from Eq. (3.4), the renormalization factor of the mass depends on the functions $\chi_n(z)$. It approaches unity as the energy levels of the perpendicular sub-bands become shallower. Thus, our results give the maximum enhancement within the weak-coupling constant scheme.

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